

On High Suppression of NO_x and CO Emissions in Gas-Turbine Plants with Combined Gas-and- Steam Cycles¹

A. A. Ivanov, A. N. Ermakov, and R. A. Shlyakhov

Abstract—In this work are given results of analyzing processes of production of nitrogen oxides (NO_x) and afterburning of CO when firing natural gas at combined-cycle gas-turbine plants. It is shown that for suppressing emissions of the said microcomponents it is necessary to lower temperature in hot local zones of the flame in which NO_x is formed, and, in so doing, to avoid chilling of cold flame zones that prevents afterburning of CO. The required lowering of the combustion temperature can be provided by combustion of mixtures of methane with steam, with high mixing uniformity that ensures the same and optimum fraction of the steam “ballast” in each microvolume of the flame. In addition to chilling, the steam ballast makes it possible to maintain a fairly high concentration of hydroxyl radicals in the flame zone as well, and this provides high burning out of fuel and reduction in carbon monoxide emissions (active steam ballast). Due to this fact the fraction of steam when firing its mixtures with methane in a gas-turbine plant can be increased up to the weight ratio 4 : 1. In this case, the concentrations of NO_x and CO in emissions can be reduced to ultra-low values (less than 3 ppm).

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INTRODUCTION

Advanced gas-turbine plants (GTP) should be located as close to power users as possible [1], because this allows reduction of capital costs and an increase in power supply reliability [2]. However, for safe location of a GTP near to a power user, especially close to a residential zone, in accordance with the government policy of environmental protection [3], it is necessary to ensure the best available environmental characteristics of these facilities by reducing the level of content of toxic components in these emissions, above all, NO_x and CO, to the minimum. This hold true, to the highest degree, for low-capacity GTP (including aeroderivative ones) in which dissipation of exhaust gases by means of high stacks is hardly practicable.

Emissions of nitrogen oxides can be considerably reduced when steam or water are supplied to the GTP circuit (Top Hat, STIG cycles, etc.); this makes it possible to increase the energy efficiency of these plants. In this case, however, increased CO content in combustion products interferes with achieving ultralow concentrations of NO_x (2–5 ppm). Injection of steam/water into the GTP circuit, while lowering the flame temperature and retarding formation of nitrogen oxides, inhibits the process of afterburning of incomplete combustion products, mainly CO. Such a method does not solve the problem of simultaneous

drastic reduction in emissions of NO_x and CO down to ultralow concentrations. The goal of our work is to determine methods for simultaneous reduction in the concentrations of these toxic components down to ultralow values on the basis of the analysis of kinetics of NO_x production and CO afterburning under conditions of GT plants with steam-and-gas cycles.

METHODS OF INVESTIGATION

Analysis of the processes of formation and suppression of toxic microcomponents in methane combustion, among other things, at supply of condensed moisture and steam, was carried out on the basis of results of mathematical simulation under conditions close to the parameters of the GTE-1500 gas-turbine plant (the Democenter of the OIVT RAN). Packages of application computer programs “neikin” [4] and “jet” [5] developed in the N.N. Semenov Institute of Chemical Physics, RAN, were used. The “neikin” package is intended for analyzing dynamics of the processes of combustion of premixed mixtures, while the “jet” package, for simulation of diffusion burning at symmetric collision of flows of the fuel and the oxidant. The scheme of methane oxidation GRI 3.0 (developed by the Gas Research Institute, Chicago) (<http://www.me.berkeley.edu/gri>) that includes 327 reactions with participation of 57 individual components forms the basis of the chemical combustion model.

Mathematical simulation of NO_x formation at turbulent burning of methane in combustors of aeroderivative GT plants having limited volume was carried out with the use of a specially constructed algorithm [6] that

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employs the formalism of joint function of probability distribution of velocity and scalars (JVS-PDF).

MECHANISM OF FORMATION AND DYNAMICS OF PRODUCTION OF NITROGEN OXIDES IN GAS-TURBINE PLANTS

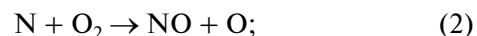
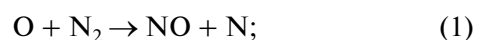
Mathematical simulation of NO_x formation at turbulent burning of methane in GT plants with the use of the JVS-PDF formalism made it possible to take adequately into account a great number of realizations of instantaneous local states of the fuel and the oxidant in the combustor and to carry out multidimensional calculation of the yield of reaction products. It was found that the correct description of dynamics of production of nitrogen oxides is only possible with due regard for local proceeding of chemical reactions in microvolumes. Rate of reaction averaged over the microvolume differs considerably (by the order and more) from the average rate determined from the average temperature. The simplest models of turbulence and combustion that do not take into account mutual effect of chemical kinetics and turbulence, in particular, those models in which the approximation of a reactor with ideal mixing is used, are unacceptable for correct determination of the yield of harmful substances, such as nitrogen oxides.

Calculations carried out according to the JVS-PDF formalism confirmed that the main contribution to emission of toxic nitrogen oxides is from thermal nitrogen oxides (Zeldovich mechanism [7]), whose formation occurs in high-temperature zones of the combustors of GT plants. Their arrangement mainly corresponds to the recirculation zone and the flame front zone of the combustor. The overwhelming bulk of nitrogen oxides is formed after completion of combustion under condition of high temperatures and at the maximum concentrations of atomic oxygen and hydroxyl radicals (OH). Arrangement of these zones corresponds, for the most part, to primary combustion of the flame extending from the fuel atomizers to the nearest holes in the flame liner through which compressed air is supplied for the flame chilling. As long as the gas mixture moves from the fuel atomizers to these holes (from the flame root to its end), i.e., as long as its high temperature is maintained, production of nitrogen oxides takes place. The concentration depends on the size of the combustor and on its hydrodynamic characteristics.

In high-temperature zones the air (oxygen)/fuel ratio is close to a stoichiometric one, and combustion takes place faster than in other zones. Excess fuel (rich mixture) or excess air (lean mixture) acts as the ballast that lowers temperature and inhibits formation of NO_x. Mixing of hot combustion products leaving the high-temperature zone with chilling excess ($\alpha > 1$) air that enters through the holes in the liner, chills the mixture sharply. This actually brings the process of NO_x formation to a halt and leads to reduction of its concentra-

tion in the gas mixture as a result of dilution of oxides that have been produced by this time. Further addition of the "cold" compressed air, entering through the next holes in the liner, to hot flow of gases dilutes combustion products even more and reduces their temperature down to the level of the working temperature (about 1000°C) of the compressor's turbine.

At maximum (local temperatures typical for combustion of mixtures of methane with air under conditions of the combustor of the GT plant with limited volume), during the residence time of a mixture in the high-temperature zone (several milliseconds), thermodynamic equilibrium as to nitrogen oxides cannot be achieved, and their concentration increases in proportion to the residence time [8]. The essence of the Zeldovich mechanism [7] of formation of thermal nitrogen oxides is that in the combustor the following reactions take place:



The first of these reactions has very high ($E \approx 75$ kcal/mole) activation energy (E) because interatomic bond (D) in nitrogen molecule is extremely strong: energy of its rupture is about 225 kcal/mole [9]. Oxygen atoms that participate in this reaction appear in dissociation of molecular oxygen—a strongly endothermic process (about 118 kcal/mole). Combination of these factors results in high sensitivity of the rate of production of nitrogen monoxide to the change in temperature. Even relatively small declines in temperature lead to considerable decrease in the rate of NO_x production. For example, lowering of temperature in the high-temperature zone of the flame by merely 100°C should result in reduction of the rate of production of nitrogen oxides by 5–6 times [8]. On the other hand, the concentration of NO is proportional to the residence time; hence, for its reduction it is necessary to shorten the time of residence of NO in the high-temperature zone as well.

Supply of "ballast" into the combustion zone for lowering temperature is a widely known technique of reducing content of nitrogen oxides in emissions from GT plants. For example, positive results were obtained when firing "lean" mixtures of methane with air in the GT plant by means of specially designed burners [10, 11]. In Fig. 1 after reference [11], it can be seen that with increasing air fraction in the "lean" mixture a decrease in the concentration of nitrogen oxides takes place. But on achieving ultra-low concentrations of NO_x (several parts per million, ppm) high incomplete burning of the fuel is observed that manifests itself in intolerable increase of content of carbon monoxide in emissions.

In STIG cycle [12] steam acts as a ballast. Due to this fact combustion of methane occurs at lowered temperatures, NO_x concentration in the combustion

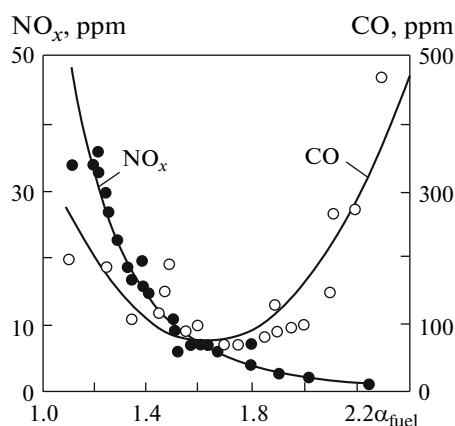


Fig. 1. Dependence of concentrations of NO_x and CO on the excess-air coefficient when combusting a preliminary prepared air-fuel mixture in the gas turbine plant.

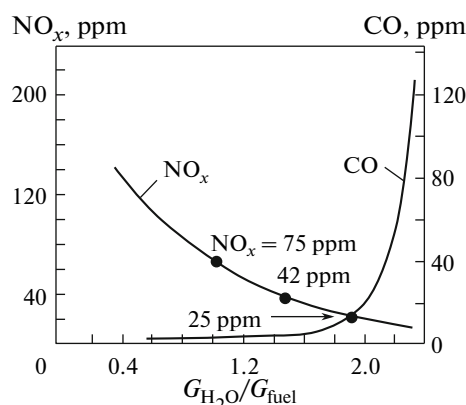


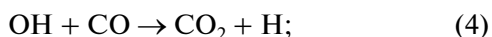
Fig. 2. Dependence of concentrations of NO_x and CO on the relative amount of steam supplied to the gas-turbine plant [13].

products decreases [13, 14]. However, with a considerable increase of the share of steam relative to that of methane, the completeness of fuel burning-out decreases also and, as a consequence, the growth of CO emissions does not allow to increase steam/fuel ratio above certain critical level (on STIG cycle this ratio is approximately 2 : 1), (see Fig. 2 [13]).

Injection of atomized water upstream of the GTU compressor has similar effect on a level of NO_x emissions [15]. This is also the case when water being injected in the forest [16] or intermediate [17] stages of the compressor to provide the so-called “wet” compression of the air. Such a scheme has been developed at IIVT RAS [18, 19]. To provide complete evaporation of micro droplets in a compressor or at the inlet to the combustion chamber, special technologies of water atomization have been developed, which make it possible to increase a share of water in the compressed air (up to 1.5 %). The further increase in the amount of water leads to inadmissible growth of carbon monoxide concentration in flue gases.

OXIDATION OF CO IN COMBUSTION OF METHANE

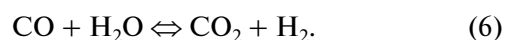
As indicated above, supply of water/steam or of air ballast (combustion of lean mixtures) results in lowering the temperature of gases, which, simultaneously with inhibited production of NO_x , is attended with an increase in the concentration of CO in emissions. Oxidation of CO takes place according to chain mechanism:



Under characteristic temperature conditions of the combustor of a GT plant reaction (5) that has activation energy $E \approx 21$ kcal/mole may turn out to be the limiting component of this process. For rapid

advancement of this reaction high concentrations of water vapor and high temperature are needed. However, at high temperatures nitrogen oxides are produced fast. Therefore, it is possible to lower temperature in the combustor for suppression of NO_x only in the relatively narrow range, until the possibility for fast oxidation of CO persists. That is what serves as a cause of a rather narrow range of relationships between flow rates of water or steam and fuel, within the limits of which it is possible to reduce simultaneously the content of both nitrogen oxides and CO in emissions from a GT plant; this rather narrow range has been found in a large number of studies.

The problem of equilibrium position in the reaction of oxidation of carbon monoxide also turns out to be essential at elevated temperatures:



At high temperatures the equilibrium (6) shifts to the left, and this leads to an increase in the CO concentration. In order for high conversion of CO into CO_2 to be achieved under such conditions, it is necessary to increase the concentration of molecules of water in local zones. Hence, supply of steam into the flame root in the combustor of the GT plant in the STIG cycle, as distinct from combustion of lean mixtures, not only leads to the lowering of the flame temperature and suppression of NO_x emissions, but also is favorable to higher afterburning of CO owing to an increase in the concentration of water in the combustion zone. Hence, steam supplied proves to be active ballast.

The impossibility of mixing uniformly in a short time the steam being injected with the flame components is the fundamental obstacle to simultaneous reduction of the concentrations of NO_x and CO in the GT plant with the STIG cycle at steam-fuel ratios exceeding critical ones. As a result, in the combustor local regions with both lowered and increased content of steam ballast relative to the average (critical) ratio

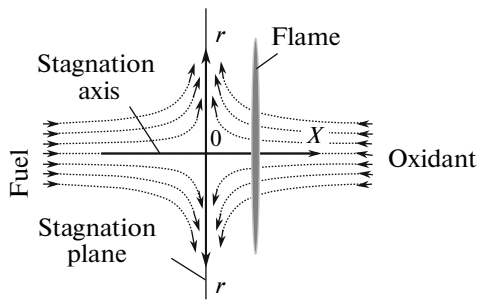


Fig. 3. The model of diffusion combustion of methane in jets moving in directions opposite each other: fuel is either methane or its mixture with steam; oxidant is air.

(2 : 1) are formed. In regions with lowered steam content a decrease in temperature is small and insufficient to inhibit considerable formation of nitrogen oxides, but in these regions CO burns out effectively. In the region with elevated steam fraction, i.e., under conditions of a drastic decrease in temperature, the process of CO burnout is unduly inhibited. Thus, the uniformity of mixing the steam ballast with the fuel affects critically the content of NO_x and CO in emissions from GT plants of gas-and-steam cycles.

COMBUSTION OF MIXTURES OF STEAM WITH METHANE IN GAS-TURBINE PLANTS

By firing premixed mixtures of methane with steam it is possible to achieve uniform distribution of steam ballast over the entire volume of the flame and provide uniform lowering of temperature in the combustor of the GT plant (see [20, 21]). In these works results are given of full-scale experiments that demonstrate the possibility of achieving simultaneously ultra-low concentrations of both NO_x and CO in emissions at steam-to-methane ratios in the mixture exceeding critical ones, up to the ratio 4 : 1.

This outcome can be explained when examining the results of mathematical simulation of diffusion combustion in flows of methane and air moving in directions opposite each other (see scheme of combustion in Fig. 3); these results were obtained by means of the "jet" package of programs. In Fig. 3 the zone of the maximum temperature (the flame) is shown by the ellipse extended in the vertical direction. On the stagnation plane the volume fractions of both methane and air are equal to 0.5 and their ratio is close to unity. But stoichiometric methane-to-air ratio (1 : 10 by volume) is realized at some distance away from this plane in the direction to the air flow. Therefore, the flame in Fig. 3 is shifted from the inhibition plane to the direction opposite to that of the air flow.

Figure 4 illustrates calculated profiles of temperature for methane-and-air flame and the flame of mixtures with the methane-to-air ratio in a mixture 1 : 1; 1 : 2, and 1 : 3; these profiles show that on threefold

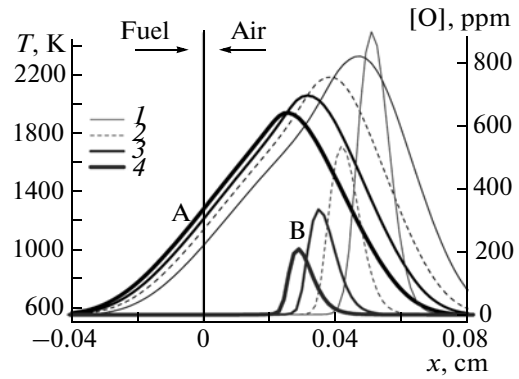


Fig. 4. Calculated profiles of temperature (A) and concentration (B) of atomic oxygen in the diffusion flame "methane + steam + air" at pressure $P_0 = 1.0$ MPa and initial temperature of the flows $T_0 = 550$ K: H₂O : CH₄ is (1) 0.1; (2) 1.1; (3) 2.1; (4) 3.1.

dilution of methane by steam the maximum of the flame temperature decreases by almost 400 K—from 2330 (without steam) to about 1930 K in its presence. In this case for the steam/methane mixture in the proportion 3 : 1, the width of the flame zone (width on the half of the height of the maximum) decreases by about 20%, while the flame itself shifts by approximately half of its width toward the fuel flow (see Fig. 4). In conformity with this, locations of the maxima of the concentrations of intermediate particles of atomic oxygen and hydroxyl radicals shift as well.

In the region located to the left of the temperature maximum the excess fuel serves as "ballast," while in the region located to the right of this maximum the excess air plays the role of "ballast." On dilution of methane by steam the fuel-to-air ratio 1 : 10, attained in the former zone of the flame without adding steam to methane, will no longer correspond to the stoichiometric methane-to-air ratio because of reduction of the methane fraction in the mixed fuel. Therefore, for mixtures of methane with steam the stoichiometric region between methane and air (the flame) moves away from the air flow in the direction of the fuel flow where the methane concentration is higher, and virtually is shifted toward the zone with lowered oxygen content.

This results in lowering the flame temperature and considerable reduction of the concentration of atomic hydrogen in it. The maximum of the concentration of atomic oxygen for each of the versions being calculated (see Fig. 4) is somewhat displaced as to the minimum of the temperature. With an increase of a steam fraction in the fuel the position of the maximum of the temperature shifts more and more toward the fuel flow where the concentration of atomic oxygen is less. Comparison of calculated profiles of the concentration of NO for the methane-air flame and for the flame with additions of steam to methane shows that the maximum (local) concentration of NO

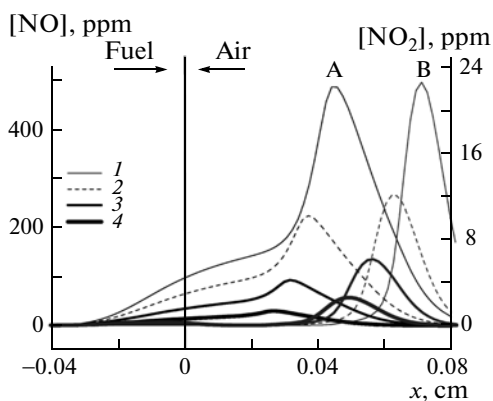


Fig. 5. Calculated profiles of concentrations of nitrogen oxide (A) and nitrogen dioxide (B) in the diffusion flame “methane + steam + air” at pressure $P_0 = 1.0$ MPa and initial temperature of the flows $T_0 = 550$ K: $H_2O : CH_4$ is (1) 0.1; (2) 1.1; (3) 2:1; (4) 3:1.

decreases from 500 ppm (without steam) to about 10 ppm on threefold dilution of methane (see Fig. 5). In this case the most drastic changes in the concentration of NO are seen from the side of the air flow. This occurs because of a shift of the flame into the region of lesser concentration of molecules, and, consequently, of atomic oxygen, where NO formation is difficult.

Nitrogen dioxide is considerably less stable than NO. Therefore, the maxima of the concentrations of NO_2 are shifted to colder zones of the flame. At a high steam fraction in the fuel one can also observe manifestation of the local maximum concentration of NO_x in the relatively cold zone from the side of the fuel flow (see Fig. 5.) However, contribution of this region to formation of NO_x is minor.

Dilution of methane by steam leads to an increase in the completeness of its combustion (the concentration of CO in emissions decreases). Calculated profiles of the concentration of CO (Fig. 6) show that a decrease in the content of carbon monoxide takes place, to the greatest degree, from the side of the air flow. The degree of reduction in the concentration of CO and positions of its maxima correlate with the profiles of the concentration of hydroxyl radicals OH, thus confirming an important role of the latter in final oxidation of CO (see reaction (4)). To attain the larger completeness of combustion of CO, the shift of the flame just toward the fuel flow, i.e., toward the higher concentration of steam, turns out to be necessary. By virtue of this, when lowering temperature in the flame zone, the concentration of hydroxyl radical decreases, but not so dramatically.

The most important condition that ensured simultaneous and high suppression of nitrogen oxides and CO in emissions from GT plants is the degree of uniformity of mixing steam with methane in the fuel mix [20–24]. The use of mixtures of natural gas with steam as fuel ensures a needed fraction of steam “ballast” in

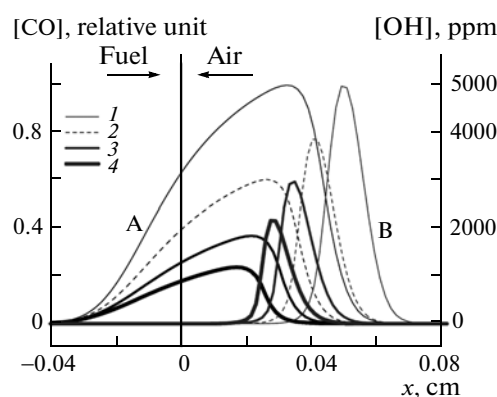


Fig. 6. Calculated profiles of concentration of carbon monoxide (A) and hydroxyl radical (B) in the diffusion flame “methane + steam + air” at pressure $P_0 = 1.0$ MPa and initial temperature of the flows $T_0 = 550$ K.

each volume of fuel. Owing to this fact, on combustion of any elementary volume of the gas mixture the temperature in the combustor decreases uniformly. Dilution of methane with steam leads to an increase in volume of the fuel mixture (by several times) as well. This levels off the ratio between volumes of fuel and air that is required for efficient and fast mixing and combustion of the stoichiometric mixture.

Mathematical simulation of the processes of combustion of steam-methane mixtures in the air showed that steam addition is the best “ballast” for suppressing emissions of NO_x and CO. As a result of steam injection, the concentration of atomic oxygen in the flame decreases, and at the same time increases the concentration of hydroxyl radicals that accelerate the combustion process and, respectively, promote reduction in CO emissions. The use of mixtures of methane with steam, prepared beforehand, as a fuel ensures the uniformity of a temperature decrease in the combustion

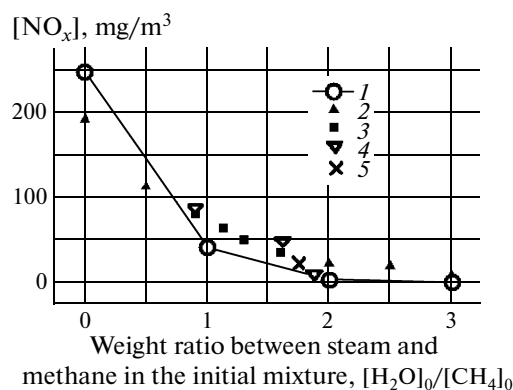


Fig. 7. Comparison between calculated values of concentration of NO_x in emissions from gas-turbine plants (1) and data obtained from laboratory (2) and full-scale tests: (3) Chevron Fr6B; (4) LM2500; (5) W501D5.

zone of the combustor of the GT plant, including the high-temperature zone (the flame).

Satisfactory agreement between the calculated concentrations of nitrogen oxides (NO_x) in emissions from GT plants at various ratios between supplied steam and methane in the mixture, and experimental data borrowed from [25] that were obtained at a number of commercially available gas turbines and in bench tests (see Fig. 7), can serve as good indirect confirmation of the correctness of the selected approaches to a description of kinetics of NO_x production and CO afterburning under characteristic conditions of GT plants with steam-and-gas cycles and the concept of their minimization considered here. Despite the fact that calculations were performed as applied to the conditions of the TV-3-117 combustor, there is very close agreement with the data for LM2500 (General Electric) and W501D5 (Westinghouse) gas turbines in which the average temperature at the combustor outlet is higher by several hundreds of degrees. Combustion in the combustor of the TV-3-117 gas turbine and in the above-mentioned GT plants made abroad is, obviously, organized in the same (and optimum) manner, and in the primary combustion flame the excess-oxidant coefficient is close to unity, and under equal initial conditions (temperature, pressure) the values of the temperature in the flame of different GT plants would be close to each other. In the primary flames of the combustor (in the first combustion area) of machines mentioned in [25], temperature is higher because of higher parameters of the compressed air (pressure and temperature), i.e., because of higher initial temperature of oxidant as compared with the TV-3-117 combustor, and, in the latter, less nitrogen oxides, while in the fore-quoted ones more of them are produced.

Hence, satisfactory agreement between calculation results and data on the concentration of nitrogen oxides observed in Fig. 7 provides evidence of correct consideration of the main determining processes and confirms the fact that the primary combustion area is the main source of NO_x. But a degree of dilution of the primary combustion flame by the "secondary" air and the processes of their mixing that are so different for the combustors of the GT plants in question obviously have influence on emissions of oxides to a considerably less degree.

CONCLUSIONS

Implementation of the gas-and-steam cycles in GT plants makes it possible not only to increase the energy efficiency but also to improve the environmental performance of these plants. Analysis of the processes of production of nitrogen oxides (NO_x) and burn off CO when combusting natural gas in GT plants showed that for simultaneous and high suppression of emissions of these toxic components it is necessary to lower the temperature in hot local regions of the flame, avoiding

in this case the chilling of its cold zones. Combustion of mixtures of methane with steam prepared beforehand with the high uniformity of mixing (deviations of the local composition from the average one should not exceed 1%) is the most promising method for solving the problem of simultaneous reduction in emissions of NO_x and CO; this offers the possibility of a considerable increase in the fraction of supplied steam in the fuel mixture, because this steam retards production of NO_x but does not prevent high burnout of CO. As calculations show, the content of NO_x and CO in emissions from a GT plant can be lowered to the level below 3 ppm, and this will make it possible to locate such plants close to power users within the limits of a city.

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